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Internal Phosphorus Loading in a Small Shallow Lake: Response after Sediment Removal

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Abstract

Mankind is taking advantage of numerous services by small shallow lakes such as drinking water supply, irrigation, and recreational function; however, many of these lakes suffer from eutrophication. Given the key role of phosphorus (P) in eutrophication process, one of the effective restoration methods especially for small shallow lakes is removal of sediments enriched with nutrients. In our study, we used interannual, seasonal, and spatial data to examine the changes in sediment P mobility after removal of sediments in 2016 from a 1-ha highly eutrophic lake. We measured the sediment redox potential, analyzed soluble reactive P (SRP) in the pore water and P fractional composition of the surface sediments, and calculated the P diffusive flux in three locations in two continuous years (2017 and 2018) after the excavation. Similar measurements were done before sediment removal at central site of the lake in 2015. Removing nutrient-rich sediment also removed 6400 kg of P, and thus the potential for release of P from sediments decreased on a long-term scale. However, a large pool of releasable P was rebuilt soon after the sediment removal due to high external P loading, resulting in extensive anoxia of sediment surface and associated internal P loading as high as 1450 mg m^{-2} $summer^{-1}$. Moreover, the Fe-P and labile P fractions were the most important sources of P release, as evidenced by their considerable seasonal and interannual changes after the sediment removal. The sediment total Fe negatively correlated with sediment P diffusive flux, pore water SRP, and near-bottom water total P and SRP concentrations which indicated a strong linkage between sediment P dynamics and Fe after the restoration. Sediment removal could be a beneficial restoration approach, but the effects on lake water quality remain only short-term unless there is an adequate control on external loading to the lake.

Keywords: eutrophication, lake restoration, phosphorus fractions, phosphorus release, sediment removal

1. Introduction

Phosphorus (P) bioavailability is regarded as one of the most important factors for determining the lake water quality in many lakes and its excessive loading causes eutrophication problems (Nicholls and Dillon, 1978; Schindler et al., 2012). The majority of P in lake ecosystems is usually stored in the bottom sediment (Pettersson, 1998). While some fractions of P in the sediment are permanently buried, others are mobile and can be recycled back into the water column. The latter include labile fraction of P and Fe bound P which can amount to more than half of the total P in the sediment (Boström et al., 1982; Søndergaard et al., 2003). Internal P cycling that reduces the water quality and delays the recovery of eutrophicated lakes originates from the mobile P pool (Søndergaard et al., 2003). Mobilization of sediment-associated P to overlaying water has usually been associated with anoxic conditions (Wetzel, 2001). When the redox potential drops below 200 mV, the ferric iron of the sediment is reduced and Fe–P complexes are breakdown. The soluble P can be further transported to the overlying lake water by diffusion due to the concentration gradient between the sediment pore water and the overlying water column (Håkanson & Jansson, 1983; Tammeorg et al., 2015).

The most common lake restoration methods that aim to improve water quality via reversing internal nutrient loading focus on increasing the retention of P (e.g., by chemical inactivation of P (Hupfer et al., 2008; Kasprzak et al., 2018) or artificial aeration and oxygenation (Beutel and Horne, 1999; Li et al., 2019). Although these techniques could be helpful for rapid restoration of cases such as lakes used for drinking water supply, they do not provide long-term solution to the problem (Horppila, 2019). Sediment removal, on the other hand, is one of most promising long-term strategies, as it permanently removes P accumulated in lake sediments (Pierce, l970; Cooke et al., 2016). Sediment removal usually has several other benefits, including lake deepening to provide habitats for fish and other biota, and thus sustaining recreational activities like fishing especially for small shallow lakes. Further benefits for the lake include

reducing nuisance growth of aquatic macrophytes (e.g., Common reed (*Phragmites australis (Cav.) Trin. ex Steud.*) and Eurasian watermilfoil (*Myriophyllum spicatum L.*); Verhofstad, and Bakker, 2019) and removing toxic sediments (Pierce, l970; Peterson, 1982). Moreover, there is a potential of recycling nutrients from excavated sediments to crop production (Canet et al., 2003; Mattei et al., 2018) or eroded coastal nourishment (De Vincenzo et al., 2019) leading to closing of nutrient cycles. Sediment removal, however, is more expensive than chemical P inactivation methods (Lembi, 2003; Bormans et al., 2016). Also, due to the potential content of toxic substances in the sediment, a strict risk assessment is needed prior to disposal or re-use of the sediment (Jeppesen et al., 2009). Further, some removal techniques such as dredging may cause disturbance to wildlife or resuspension of sediments and release of toxic substances to overlaying water (Jeppesen et al., 2009; Bormans et al., 2016).

Most recently, Lürling et al. (2020) have reviewed the effects of sediment removal on internal P loading and water quality in a large number of lakes varying in size worldwide. Generally, the removal of sediment has in many cases reduced the internal P loading immediately and substantially. The positive effects for lake water quality included decline in water column nutrients, reduction of phytoplankton, disappearance of cyanobacterial blooms, and increased coverage of macrophytes. However, for some cases, although the internal loading of P was reduced by the sediment removal, long-term success has been hindered by a continuously too high external loading (Björk et al., 2010; Bormans et al., 2016) or due to insufficient removal of surface sediment (Liu et al., 2015; Phillips et al., 2015). Hence, the sediment removal projects designed to restore eutrophic lakes showed mixed results. Moreover, there are just a handful of studies available exploring the effects of full removal of all the lake sediment on P internal loading and lake water quality. In the current study, we monitored internal P loading in a small lake (about 1 ha) after the full removal of sediments. The comprehensive

assessment of internal P loading in the lake by monitoring spatial and temporal variations will improve understanding of the key processes for the success of restoration.

2. Materials and Methods

2.1. Study site

The study was conducted on the man-made Lake Mustijärv (created in around 1985, surface area 1 ha) located on Kurika stream about 1 km west of Viljandi, Estonia (58°21'55.8"N 25°32'32.6"E, normal water level at 65 m above sea level, Fig. 1) in 2015–2018. Based on daily meteorological data at Viljandi meteorological station located at a distance of 3.6 km from the lake, the mean air temperature of the growing period (May to October) was 12.2 and 15.0 ˚C in 2017 and 2018, respectively. Also, the mean precipitation was 80.4 in 2017 and 67.6 mm in 2018, respectively. The growing period was drier and hotter in 2018 in comparison with both 2017 and long-term 1981–2010 mean (12.8 ˚C and 76.2 mm) in Viljandi (EWS, 2019).

The majority of nutrients were carried into the lake by the stream from the east side of the lake (Fig. 1a). The shore of the lake was covered by sandy loam soil (sand 60%, silt 27% and clay 13%) and the lake was rich in loamy sediment (sand 40%, silt 42% and clay 18%) before excavation. By 2015, the lake was heavily eutrophicated (Table 1, Fig. 1a), even to the extent that willows grew on the silt piles in addition to widespread macrophytes including mainly Bulrush (*Typha latifolia L.*) and Common reed. The lake had only 0.3 ha free-water surface with a mean depth of 1.1 m and maximum depth of 1.6 m at that point (Figs. 1a and 1d). In 2016–2017, the lake was fully desilted to the lakebed via excavation of the whole sediment (1.1 m depth on average) to a mean lake depth of 2.2 m and maximum depth of 3.7 m (Figs. 1b and 1d). Also, two deeper sediment collection basins were introduced, one close to the inflow and another at the center of the lake (Fig. 1c). Further, an island of rocks was introduced in the center of the lake, with the aim of being the spawning habitat for fish. Further, the downstream part of the lake was expanded by excavating new basin to the lake so that in spring

2017, after the lake restoration was finished, the Lake Mustijärv had a surface area of 1 ha, length of 257 m, and the largest width of 58 m (Fig. 1c). The lake morphometry was mapped by probing the geolocated sites in 2013 and in 2018 (Fig. 1d).

2.2. Data collection and analyses

Sediment P mobility was characterized by measuring the redox potential and pore water concentrations of SRP and dissolved Fe in surficial sediments (uppermost 3 cm layer), calculation of SRP diffusive flux, and analyzing the composition (P fractions, organic matter and Fe content) of surficial sediments. For this purpose, three sampling locations were selected in the lake inflow (IF), center (CE), and outflow (OF) areas (Fig. 1c). Sampling was carried out at each sampling location in August 2017 and in June, August, and October 2018. During each sampling campaign, environmental variables potentially affecting the biological, chemical, and physical transformations at the sediment-water interface (i.e., temperature (T), pH, dissolved oxygen (DO), and electrical specific conductance (SPC)) were measured in-situ with a handheld multi-parameter water quality analyzer (YSI Professional Plus, 6600V2, Yellow Springs, Ohio, USA). Water samples from the surface water layer (0.5 m from the lake surface) and water layer overlying the lake bottom (0.5 m above the lake bottom) were taken with a Limnos sampler. From lake water samples, the concentrations of the parameters potentially important for the phosphorus recycling were determined including SRP, TP, total Fe and dissolved Fe. Sediments were sampled with an HTH gravity corer (Renberg and Hansson, 2008). Similar sampling of sediment and lake water was done in November 2015 at the deepest location of the lake, the only spot without macrophytes which enabled us to describe situation before sediment removal.

For redox potential measurements, the sediments withdrawn with the HTH gravity corer were subsampled (two sub-samples) into the plastic tubes (inner diameter = 3.5 cm, height = 14 cm) so that 2/3 of the tube volume was filled with sediments covered by water (1/3 of the tube volume). The tubes were sealed with caps and transferred to the laboratory in the thermo-isolated box. Immediately after

sampling, the redox potential of the sediment was measured directly in the tube down to the 3 cm below the sediment surface in 1-mm steps using a redox microelectrode (Unisense RD100 microsensor, Aarhus, Denmark) and a Ag/AgCl reference electrode. The measurements were repeated three times for each tube (Fig. 3). Prior to measurements a two-point calibration was performed in saturated quinhydrone buffer solutions with redox potential of respectively 385 mV and 486 mV at 10 ºC.

At each sampling location, three sediment cores were concurrently obtained by the HTH gravity corer for pore water extraction. Pore water was extracted using Rhizon Soil Moisture Sampler (Rhizosphere Research Products, Wageningen, the Netherlands) inserted horizontally at depths of 1, 2, and 3 cm (Fig. 4). From the pore water, SRP (ISO 15681-2) and Fe concentration (ENS-EN ISO 17294-2) was also determined. The diffusive SRP flux (mg P m^{-2} day^{-1}) was calculated according to Fick's first law of diffusion (Berner, 1980):

$$J = \Phi \times D_s \times \frac{\Delta C_{SRP}}{\Delta Z} \quad (1)$$

Where ϕ is the sediment porosity, D_s (cm^2 s^{-1}) the diffusion coefficient of phosphates, and $\Delta C_{SRP}/\Delta Z$ the concentration gradient between the sediment pore water and the overlying water column. In the calculation of concentration gradient, the SRP concentration of pore water in the 1-cm surface sediments (ΔZ = 0.5 cm) and the SRP concentration of the overlying lake water were used. The porosity value for the uppermost 3 cm of the surface sediment was used. The diffusion coefficient for SRP at 25 °C (D25 °C) in sediment-water systems is $6.12 * 10^{-6}$ cm^2 s^{-1} (Yuan-Hui and Gregory, 1974). The temperature dependence of the D25 °C was taken into account according to the Stokes-Einstein relation (Lewandowski and Hupfer, 2005):

$$D_i = D_{i,25℃} \times \frac{\nu_{25℃} \times T}{\nu_T \times T_{25℃}} \quad (2)$$

where T is the temperature during sampling in Kelvins, $T_{25°C}$ is the absolute temperature at 25 °C (298.15 K), $\upsilon_{25°C}$ is the dynamic viscosity of water at 25 °C (0.8903 g m^{-1} s^{-1}), and υ_T the dynamic viscosity of water at a given temperature T (g m^{-1} s^{-1}).

Additionally, 3 cm of the uppermost surface sediments were sampled to analyze P fractions, concentration of Fe, and organic content of the sediments. The P fractionation was conducted following the modified Williams protocol by Ruban et al. 1999, because the method was proved to be the most appropriate in achieving comparability in interlaboratory study (Ruban et al., 2001). The studied P pools included the total P (TP), organic P (OP), inorganic P (In-P), P bound to Al and Fe (hydro) oxides (targeted fraction is the Fe-bound P, i.e., Fe-P), and P bound to Ca (Ca-P). To solubilize Fe and Ca, 1 mol L^{-1} NaOH and 1 mol L^{-1} HCl were used, sequentially in the same aliquot (0.2 g of dried sample). With another aliquot, total P was extracted with 3.5 mol L^{-1} HCl. Using the third sediment aliquot, the In-P was extracted by 1 mol L^{-1} HCl and the residual was treated at 450 °C to analyze OP. Additionally, labile P (Plab) was extracted with 1 mol L^{-1} NaH_4Cl as a part of Hieltjes–Lijklema (1980) protocol due to the importance of bioavailability of Plab for the lake water quality. In the laboratory, the sediment samples were dried at 105 °C for approximately 24 h to obtain their dry weights. The organic matter of sediments was determined by loss-on-ignition at 550 °C for 2 h. Fe in sediment samples was analyzed by inductively coupled plasma-optical emission spectroscopy (ICP-OES; Thermo-Fisher iCAP3600 MFCDuo, Thermo Fisher Scientific, Cambridge, UK). From surface and bottom water samples, TP, SRP (ISO 15681-2), total Fe, and dissolved Fe concentrations (EVS-EN ISO 11885) were determined.

Finally, internal P loading from anoxic area in summer 2018 was calculated as a product of anoxic factor (AF) and the release rate (RR) of P from sediments (Nürnberg, 1984):

$$Summer\ internal\ P\ loading = RR \times AF \quad (3)$$

The mean P diffusive flux (mg m^{-2} day^{-1}) from June to August was used as a measure of P release rate (RR). AF was calculated by multiplying the percentage of anoxic areas and duration of anoxia. Oxygen concentration measurements in the lake water overlying lake bottom since June indicated anoxia (DO concentrations less than 2 mg/l) at lake depths from 1.5 m, suggesting that almost entire area of the lake was involved in P release due to anoxia. No anoxia was observed by the end of September, which results in the approximate duration of anoxic period of 92 days. Alternatively, we also calculated the internal loading using a modelled AF as defined by Nürnberg, 1996:

$$AF_{pred} = -36.2 + 50.2\log(TP_{summer}) + 0.762\frac{Z}{A^{0.5}} \quad (4)$$

where TP_{summer} is average summer TP in lake water (µg l^{-1}), Z is lake mean depth (m), and A is lake surface area (km^2). Epilimnetic TP values from June to August were averaged as TP_{summer}.

2.3. Statistical analyses

Statistical analysis was performed using R v3.5.3 software. Using Levene's and Shapiro-Wilk's tests, Lake Mustijärv data were tested and passed the tests of normality and homogeneity of variances. The interannual (2015, 2017, and 2018), seasonal (June, August, and October), and spatial (locations IF, CE, and OF) variations for lake sediment and water properties were analyzed with ANOVA. When there was a significant effect in the ANOVA models ($P < 0.05$), Tukey HSD tests were subsequently run to identify significant differences between the means. The correlations between the lake sediment and water parameters were studied by Pearson's analysis.

3. Results

3.1. Interannual variations in sediment P mobility

3.1.1. Changes in mobility of P before and after sediment removal, autumnal data (Nov 2015 and Oct 2018)

Removal of approximately 7500 m^3 sediment (1.1 m excavation depth) from Lake Mustijärv resulted in the removal of about 6400 kg of the total phosphorus from the lake and increasing the volume of the lake to ~ 21000 m^3. In 2017–2018, new sediment materials entered the lake (Fig. 1e), consisting of a) the surface runoff from sediment drying on lake shores before establishment of grass cover in 2018 and b) sediment material carried downstream from the cleaning processes of Kurika stream banks (major flux, Fig. 1c). The total P concentration in surface sediment in 2018 was 60% of that in 2015 (Fig. 2). In 2015, the dominant phosphorus fraction in Lake Mustijärv was the Ca bound fraction (42%), i.e., phosphorus that is usually considered biologically inaccessible in practice. However, the total share of the fractions with the highest potential of biological availability (Plab and Fe-P) was high as well (37%). After the removal of sediment, the share of Fe-P and Ca-P decreased (~ 6%), and OP contributed 18% more in the total P pool of surface sediment (Fig. 2). Additionally, the sediment Fe concentration was 20% lower in 2018 and Fe/P ratio increased from 10.4 to 14.3. The organic matter content of the surface sediment was significantly higher in 2015 (31%) than in years after lake excavation (22% as a mean for 2017 and 2018, Table 2).

Redox potential at CE loation was close to 200 mV at the sediment-water interface in 2015 and 2018 (Fig. 3), indicating similarly high risk for P mobilization. The vertical changes in the redox potential values were very similar when comparing those before the excavation in 2015 and after the excavation in 2018. They had a rapid decrease down to a depth of 1 cm, where the values were 153 (±21) and 149 (±10) mV, respectively and remained below these values in the deeper layers. Sediment pore water SRP concentration at

the depth of 1 cm did not differ significantly between the studied years while the SRP concentration was two times lower at the depth of 3 cm in 2018 than in 2015 ($P < 0.05$; Fig. 4). Diffusive flux at CE location in 2015 (4.37 mg P m^{-2} day^{-1}) was about 28% of that in 2018, which may be partially explained by the higher temperatures during sampling in autumn 2018 (October 2018) compared with autumn 2015 (November 2015). Noteworthy, although the calculated diffusive fluxes were higher after the excavation, the total P pool of sediments, and thus the potential for internal loading decreased on the long-term scale.

3.1.2. Changes in mobility of P in two subsequent summers after sediment removal (August data of 2017 and 2018)

When focusing on years after lake excavation, a significant increase in the concentration of sediment labile P was observed between August 2017 and August 2018 as a mean of three locations (10 vs. 26 mg kg^{-1}; $P < 0.05$). Also, a slight statistically insignificant increase was observed in the sediment total P concentration and proportion of Fe-P in the sediment P pool (Table 2; Fig. 2).

Comparing the redox profiles one year and two years after the sediment removal revealed that the redox potential of surface sediment was generally higher in August 2018 than in August 2017 (Fig. 3). As a mean of all three locations, the pore water SRP concentration in the surface sediment was 1.7 times greater in two years after sediment removal (2018) than 2017 (Fig. 4). Similarly, the mean diffusive flux of SRP was 1.6 times higher than in 2017 (Table 2). In details, the redox potential in 2017 showed a rapid decrease down to the depth of 1 cm at IF and OF locations, where the values declined from 229 to 116 mV at IF and from 305 to 257 mV at OF. In August 2018, the redox values of all three locations were significantly greater and the vertical changes were smaller than in August 2017. Similar to redox potential, the SRP concentration at surface sediment significantly differed between August 2017 and 2018 at both IF and OF locations with 3 to 5 times greater values in 2018, but the difference was not significant at CE location.

Regarding the properties of water overlying lake bottom, DO, pH, SPC, and temperature were lower in 2018 than 2017 (Table 2). DO was 4.7 mg l^{-1} in 2017 and 0.7 mg l^{-1} in 2018. In addition, the concentrations of total P, SRP, and total Fe in near-bottom water were greater in 2018 than 2017.

3.2. Spatial variation in sediment P mobility in Lake Mustijärv after lake excavation

The total P concentration and all P fractions of sediment had considerable spatial variation in Lake Mustijärv with a decreasing trend from IF to CE and then to OF location when comparing the mean values of August 2017 and August 2018 data (Fig. 1c and Fig. 2). The trend was reverse for the sediment Fe concentration and Fe/P ratio (Table 2). The OF location where was the expanded area of the lake after excavation, differed significantly from the other two locations with almost 50% lower P content (Fig. 2) in sediment and two-fold Fe/P ratio ($P < 0.001$), while the IF and CE locations were not significantly different from each other in these regards. Also, at OF location, the majority of TP was Ca-P (52%) and the Fe-P comprised a significantly smaller proportion of the total P pool of the sediment compared to IF and CE locations (19% vs. 32%). The organic matter content of sediment material significantly decreased from 28% at IF location to 22% at CE and to 12% at OF. Also, the IF location had 21% lower LOI value in August 2018 than August 2017 ($P < 0.001$) but there were no significant differences between these years at CE and OF locations (Table 2).

In general, the IF and OF locations had greater vertical changes in redox potential profile during the years after excavation while less pronounced changes were detected at CE (Fig. 3). As a mean of values in 2017 and 2018, the pore water SRP concentration had the lowest value at OF which were significantly different from CE and IF locations (Fig. 4). Also, the mean value of diffusive flux varied spatially among three locations (Table 2). The lowest value of SRP diffusive flux was observed at OF (0.83 mg P m^{-2} day^{-1}) which was only 4% of the flux at CE and IF locations ($P < 0.001$). In details, the diffusive flux in CE (34.6 mg P m^{-2} day^{-1}) was significantly greater

than in IF (8.0 mg P m^{-2} day^{-1}) and IF had higher flux than OF location (0.25 mg P m^{-2} day^{-1}) in 2017 ($P < 0.001$). Similarly, in 2018, OF showed significantly lower diffusive flux than IF and CE. In addition, negative flux implying that sediment served as a sink of P were only observed at OF location in October 2018.

3.3. Seasonal variation of P loading in Lake Mustijärv in 2018

In August 2018, the concentration of TP at IF location was 0.28 mg l^{-1} in both lake surface and near-bottom water which was more than two-fold than those in June and October (Table 2). The average depth of the newly formed sediment was 0.19±0.12 m (Fig. 1e) with the highest depth of sediment in the accumulation basins at IF and CE locations.

Some of the phosphorus forms in surficial sediment showed marked seasonal variations during 2018 (Fig. 2). The sediment total P declined from June (1500 mg kg^{-1}) to August (1333 mg kg^{-1}) but had 11% increase from August to October. The labile P and Fe-P fractions had significant seasonal changes with lowest contribution to sediment TP in June ($P < 0.05$) while their share was highest in August (33%). On the other hand, the share of Ca-P in sediment P (45%) and the Fe/P ratio with the value of 17 were seasonally stable and the contribution of OP to the total P decreased from June to October. The content of sediment organic matter had a downtrend from June to August but significantly increased from August to October at IF and CE locations (Table 2). No significant seasonal changes were observed at OF.

During three studied months, the redox potential at the sediment water interface was close to 200 mV at IF and CE locations, and somewhat higher (around 300 mV) at the OF location (Fig. 3). The decline in redox potential with depth varied depending on month at locations IF and OF, while CE did not show any pronounced seasonal changes. In general, IF and OF locations had the greatest vertical changes in redox potential in October, when the mean value declined around two times. At OF location, the profile of redox potential in

October resembled that in June, while interestingly, the critical value for the release of P (200 mV) was not reached within the top 3 cm of sediments in August. In IF, a similar pattern for redox potential was observed in August and June, and the reduction trend of redox values were clearly less pronounced than in October. The mean value of pore water SRP concentration in 1 cm sediment layer in August was two-fold of that in June and October at CE ($P < 0.05$; Fig. 4) and IF locations. However, SRP values at the sediment depth of 3 cm showed no significant seasonal changes in these locations. In OF, the mean SRP values in surface sediment were significantly greater in August than in June and October. Also, the SRP diffusive flux showed considerable seasonal variations in 2018 (Table 2). The highest values of flux at CE were observed in August (30 mg P m^{-2} day^{-1}) which was 43% greater than in June and 2.5 times higher than the flux in October ($P < 0.05$). The trend was the same for IF but was not statistically significant due to large variation. In OF, the diffusive flux with negative value of -0.03 mg P m^{-2} day^{-1} in October was significantly lower than in June and August. The internal P loading in Lake Mustijärv was estimated to be 1450 mg m^{-2} $summer^{-1}$ as the mean for the whole lake. Internal loading of P based on the equation of modelled AF resulted in a value of similar level, 1566 mg m^{-2} $summer^{-1}$, for the summer 2018.

3.4. Correlation analyses among lake water and sediment properties

A significant positive correlation was found between the diffusive flux (J) and the following parameters (Fig. 5): surface water total P concentration(s TP), bottom water specific conductance (SPC_b), SRP (SRP_b), total Fe (TFe_b), and dissolved Fe concentrations (DFe_b), and sediment labile P (Plab), Fe bound P (FeP), organic P (OP), and pore water Fe concentrations at top 3 cm of sediment core (Fe1, Fe2, and Fe3). Additionally, diffusive flux correlated negatively with near-bottom water dissolved oxygen (DO_b) and pH (pH_b), sediment Fe concentration (s Fe), and Fe/P ratio (Fe.P; Fig. 5). Although all sediment P fractions showed significant negative correlation with Fe/P ratio, the OP and Fe-P showed the strongest correlation. Also, Fe/P ratio negatively correlated with LOI and TFe_b ($P < 0.05$).

Among all P fractions, the labile form of P (Plab) and Fe-P were the only fractions which positively correlated with SRP diffusive flux, pore water SRP at top 3 cm of sediment core, and pore water Fe2 and Fe3 concentrations ($P < 0.05$; Fig. 5). Moreover, bottom water pH had significant negative correlation with J, pore water Fe1, SRP1, and SRP2.

4. Discussion

After sediment removal in Lake Mustijärv, the TP pool decreased, and thus the potential for P release from sediments decreased in long-term. Still extensive anoxia of surficial sediments and water overlying lake bottom and high P release by diffusion were observed in first years following the excavation. The values of diffusive flux of P measured after sediment removal were in the range of the P release rates under anoxic conditions in hypertrophic lakes (Nürnberg, 1984; Carter and Dzialowski 2012). Despite the sediment removal, lake trophy remained high, which was evidenced also by the biota composition studies in this lake. The TP concentration in the surface water layer was far above 100 µg/l, which is considered as a concentration level between eutrophic and hypereutrophic lakes (Nürnberg, 1996). A pool for internal release of P was rebuilt soon after sediment removal, which was not a surprise given that the external P loading exceeded the critical level (as defined by Vollenweider, 1978) nine-fold (unpublished data). Similarly, there are many examples of lakes worldwide, in which measures targeting lake water improvement via reduction of internal P loading were unsuccessful due to the high external P supply (Cooke, 2005; Bormans et al., 2017). As an example, the sediment removal from the shallow Lake Brabrand in Denmark led to an increase in water depth and a reduction of phosphorus release from the lake bottom. The lake remained, however, in the turbid state because the external nutrient loading has not been reduced sufficiently (Søndergaard et al., 2000).

The internal P recycling sustained eutrophication in Lake Mustijärv. We observed TP increases during summer when a trough of the hydrograph appears, a well-documented phenomenon increasing internal P loading in shallow lakes (Nürnberg, 2009, Kowalczewska-Madura et. al., 2015). Moreover, diffusive flux of P exhibited a seasonal pattern mimicking the seasonal variation in lake water phosphorus concentration. Shallow lakes are particularly vulnerable to internal loading, because these lakes are frequently mixed to the bottom and sediment-released phosphorus readily enters the trophogenic zone and trigger the high primary production (Welch and

Cooke 2005; Søndergaard et al., 2003). Moreover, there is often lack of direct evidence for anoxic P release from bottom sediments in shallow lakes (Nürnberg, 2009).

Of the main components of TP (Plab, Fe-P, Ca-P, and OP), potentially algal-available P (fractions Fe-P and Plab) notably had seasonal changes and became the most important contributors to sediment P release. The proportion of labile P and Fe-P increased significantly by August being a source of dramatic increase in SRP concentrations in water overlying lake bottom. Moreover, strong correlations of Fe-P and labile P with the diffusive flux ($r > 0.68$, $P < 0.05$) suggested that they were the most mobile P fractions cycling between sediment and water seasonally. Fe-bound P is easily released into interstitial water at reduced conditions of surface sediments like we observed in Lake Mustijärv. This finding is consistent with other studies reporting that the Fe-P is the primary contributor to internal P loading and the most important algal-available P fraction in the highly eutrophic shallow lakes (Petticrew and Arocena, 2001; Zhu et al., 2013). Our study highlights the clear linkage between sediment P and Fe dynamics after the sediment removal which was further supported by the strong negative correlation of total Fe in surface sediment with the pore water SRP, dissolved Fe, and bottom water TP and SRP concentrations (Fig. 5). In addition, the sediment Fe/P ratio, indicating sediments ability to bind P (Jensen et al., 1992; Caraco et al.,1993; Søndergaard et al., 2001), had a strong negative correlation with the diffusive flux of P (Fig. 5). Similarly, numerous other studies recognized that the intensity of phosphorus loading from bottom sediments can be determined by the contribution of particular P fractions to the TP content (Søndergaard et al., 1996; Kowalczewska-Madura et al., 2015) as the biological accessibility of the fractions differ. Examining the proportion of each fraction provides important information on the stability of P forms in sediments and their potential to be released to the water column (Kaiserli et al., 2002). For example, Jing et al. 2015 noted that monitoring of Fe cycle in lakes after sediment dredging helps us to understand the environmental effects of restoration process.

Changes in the concentration of Fe bound P could most likely explain also interannual variations in the release of P from sediments. In 2018, i.e., two years after sediment removal, the diffusive flux of P was generally higher than in 2017. At the same time, the Fe-P fraction, which represents potentially bioavailable P, constituted larger amount of TP in 2018 than in 2017. Apparently, there was an increase in external P loading in 2018 to Lake Mustijärv, because Fe-P has been often shown to be of anthropogenic origin (Ruban et al., 2001) and inflow of nutrients from the catchment can be one of the main contributors of Fe-P content in sediment (Gao et al., 2005; Zhu et al., 2013). Also, over the entire lake, IF sampling location (i.e., in the close vicinity to the upstream) had the greatest share of Fe-P in the sediment TP pool and significantly higher P flux in August 2018 than August 2017. Considering the idea that external P loading is the main contributor of Fe-P content in sediment, nutrient input via the inflowing stream with TP concentration of 220 µg/l (measured in autumn 2018 during monitoring of stream, unpublished data) exceeding more than 3 times the level before sediment removal (Table 1) could be the reason of higher P flux in 2018 particularly at IF. In the OF, however, the total P pool was lowest, and the effect of external loading was greatly reduced as the Fe-P only took up to 19 % which could be due to the sediment accumulation basins across the lake where external nutrients were already captured (Fig. 1e). In addition to the external loading, the significant higher temperature during sampling in 2018 than in 2017 may partially explain the higher P flux in 2018.

The OP content in surface sediments showed significant positive correlations with P flux and SRP in pore water of upper 3cm of sediment, suggesting also this fraction as a potential source for the sediment release of P. OP that strongly positively correlated with LOI value (r= 0.87), is complicated in composition and structure, but it has been shown that half of OP components can be converted into algal-available P by mineralization (Rydin, 2000). In October, at lower water temperatures the organic matter production decreased and there was no anoxia in the water layer overlaying lake bottom. Although, the calculated diffusive flux of P at IF and CE (> 12 mg P m^{-2}

d^{-1}) was comparable to those in June, the actual benthic fluxes were most likely lower, since the oxic sediment surface layer in autumn retained phosphorus effectively. The dynamic ratio (i.e., the square root of the lake surface area in square kilometers divided by its mean depth in meters; Håkanson, 1982) of Lake Mustijärv was lower than 0.1. It is an indication that the whole lake bottom dynamics is dominated by slope processes, erosion, and transport processes of the sediment material. Moreover, there was a decreasing trend of LOI value from IF to CE and to OF (Table 2) indicating that the allochtonous organic materials, associated with increased external load, settled on the lake bottom mainly before the OF location. Moreover, currents may be stronger close to the outlet, and thus settling of light organic particles is lower.

There were no seasonal changes in Ca-P while its portion in TP pool decreased after sediment removal. Ca-P tends to be from weathering and erosion origin with little risk of release (Ruban et al., 1999). Also, it became evident that the OF location had lower portion of potentially releasable P (Fe-P and Plab) and Ca-P was the major contributor in its TP pool (52%) which was significantly greater than that of IF and CE locations. This agreed with notably lower P release from sediments at OF location.

The highest sediment P release occurred at the deepest location of the lake. The wide variations of diffusive flux in this site emphasize notable estimates of internal P loading due to anoxia likely caused by high rates of organic matter decomposition. Moreover, there was also a clear temperature gradient during some of the sampling times (Table 1), which enhances the anoxia (or enables it by causing stagnation) caused microbial degradation. Large nutrient loading brought from upstream stimulates the high primary production resulting in oxygen depletion and recycling of P back to the water column. Nutrient inputs by streams are considered as point sources nutrient inputs which enter a lake at one location rather than spread over the whole lake (Janssen et al., 2019). Furthermore, in lakes with low value of dynamic ratio (< 0.8), sediments are concentrated in central deeper areas (Bachmann et al., 2000). Sediments tend to move

until they end up in an undisturbed area where they can accumulate. If such areas do not exist, the sediments will continue to resuspended and will be widely distributed over the lakebed (Bachmann et al., 2000). Entrapment of nutrient-rich sediments already in the inflow of the lake leads to a decrease in primary production towards the outflow as we observed in Lake Mustijärv (Table 2, Fig. 1e). This inference directs us to suggest that introducing more accumulation basins may help to entrap the point source nutrient inputs more efficiently. More research is needed to explore to what extent such accumulation basins, if emptied regularly, can help to localize the high-P sediment caused issues and facilitate improvements in water quality in majority of lake area.

5. Conclusions

Our study used the interannual, seasonal, and spatial data of Lake Mustijärv to examine the factors behind sediment P release from a small shallow lake after sediment excavation. Sediment removal decreased the total P pool, and thus a potential for the release of P from sediments on the long-term scale. Still high rate of summer P release was observed in a few years after sediment removal. High pool of releasable P was rebuilt soon due to high external P loading, resulting in extensive anoxia of sediment surface and associated internal P loading as high as 1.4 g m^{-2} $summer^{-1}$. In our study, the P release was clearly linked to the Fe-P concentrations of sediments and coupled to oxygen depletion of near-bottom water. Moreover, the Fe-P and labile P with the highest mobility between sediment and water Showed considerable seasonal and interannual changes after sediment removal and became the most important source of summer P release in Lake Mustijärv. The spatial data revealed that the deepest area of the lake had the major contribution on the phosphorus loading due to anoxia. The outflow location where was the expanded and shallowest area of the lake after excavation, had the lowest amount of potentially releasable P and thus, the lowest flux of P from sediment to the water column. This could be due to the trapping of the external nutrients by accumulation basins before reaching this area. This suggests that introducing more accumulation basins close to the upstream may help to entrap point source nutrient inputs more efficiently before reaching to the central location of the lake.

Sediment removal as a lake restoration technique may have a range of benefits including reduction of internal P loading which reduces algal biomasses and increases water clarity. Moreover, removal of P-rich sediment opens the opportunity to re-use of P in a sustainable manner. Nevertheless, any lake restoration method may not be successful without a sufficient control of external loading.

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Figure 1. a) Aerial photo of eutrophicated Lake Mustijärv located west of Viljandi, Estonia and sampling locations in 2015. b) Sediment excavation of the lake in 2016. c) Sampling locations in 2017 and 2018 (yellow points). d) Depth of water in 2015 and 2018. e) Depth of newly formed sediment measured in 2018.

Figure 2. Sediment total phosphorus concentration (TP), organic P (OP), inorganic P (In-P), Fe bound P (Fe-P), Ca bound P (Ca-P), labile P (Plab) concentrations, and their contribution in sediment TP pool (shown as % values of TP pool above the bars) from 2015 to 2018 in locations close to the inflow (IF), center (CE), and outflow (OF) of Lake Mustijärv (Center was the only station available for measurements in 2015).

Figure 3. Redox potential (mean as a solid line, standard deviation as shaded area) of the uppermost 3 cm of surface sediments in locations close to the inflow (IF), center (CE), and outflow (OF) of Lake Mustjärv from 2015 to 2018 (Center was the only station available for measurements in 2015). The critical value of 200 mV is shown as a vertical black line.

Figure 4. Mean soluble reactive phosphorus (SRP) and dissolved iron (Fe) concentrations in the sediment pore water separated from the uppermost 3 cm of surface sediments in locations close to the inflow (IF), center (CE), and outflow (OF) of Lake Mustijärv. The error bars are the standard deviations.

Figure 5. Correlation network based on Pearson correlation coefficients among lake sediment and water properties in Lake Mustijärv. The black lines indicate positive correlations; the red lines indicate negative correlations. The thickness of the line shows the strengthen of the correlation. Only significant correlations were shown (cutoff value: 0.55). Also, correlations between variables with high collinearly (dependency) were not shown. The abbreviations for the variables are translated as follows: SRP diffusive flux (J), sediment organic matter (LOI), sediment total P (sTP), organic P (OP), inorganic P (inP), Fe bound P (FeP), Ca bound P (CaP), labile P (Plab), sediment Fe (sFe) concentrations, Fe/P ratio (Fe.P), pore water Fe concentrations at top 3 cm of sediment core (Fe1, Fe2, and Fe3), SRP at top of 3 cm of the core (SRP1, SRP2, and SRP3), redox potential values at 5 and 25 mm of sediment core (Rdx5 and Rdx25), bottom water total P (TPb), soluble reactive P (SRPb), total Fe (TFeb), dissolved Fe (DFeb), dissolved oxygen (DOb), pH (pHb), specific conductance (SPCb), temperature (Tb), and surface water total P (TPs), soluble reactive P (SRPs), total Fe (TFes), dissolved Fe (DFes), dissolved oxygen (DOs), pH (pHs), specific conductance (SPCs), temperature (Ts).

Table 1. The water properties in the middle of the lake, upstream and downstream of Lake Mustijärv before excavation in November 2015

Parameter	2015		
	Center	Upstream	Downstream
TN	–	1.1	1.1
TP	0.044	0.06	0.044
$P0_4^{3-}$–P	0.035	0.044	0.037
BOD	–	1.5	1.9
DO	–	9.9	13
SS	–	5	3.5
pH	–	8.1	8.1

TN: total nitrogen (mg/l); TP: total phosphorus (mg/l); $P0_4^{3-}$ -P: phosphate (mg P/l); BOD: Biochemical oxygen demand (mg O_2/l); DO: dissolved oxygen (mg/l); SS: suspended solids (mg/l).

Table 2. The values of concentration of dissolved oxygen (DO), electrical specific conductance (SPC), pH, temperature (T), total P (TP), soluble reactive phosphorus (SRP), and total Fe (TFe) in the surface and near-bottom water, and sediment pore water SRP at 1-cm depth, Fe concentration, Fe/P ratio, LOI value, and diffusive fluxes of SRP (J) in Lake Mustijärv from 2015 to 2018. The values in parenthesis are the mean standard error.

Sampling	surface water							near-bottom water							sediment				J
	DO	SPC	pH	T	TP	SRP	TFe	DO	SPC	pH	T	TP	SRP	TFe	SRP	Fe	Fe/P	LOI	
	mg l^{-1}	µS cm^{-1}		°C	mg l^{-1}	mg P l^{-1}	mg l^{-1}	mg l^{-1}	µS cm^{-1}		°C	mg l^{-1}	mg P l^{-1}	mg l^{-1}	mg P l^{-1}	g kg^{-1}		%	mg $m^{-2}d^{-1}$
2015-Nov.																			
CE	–	–	–	–	–	–	–	–	–	–	1.50	0.044	0.035	0.23	1.0 (0.10)	25	10.4	30.7 (0.2)	4.37 (0.4)
2017-Aug.																			
IF	8.41	501	8.36	19.6	–	–	–	7.91	505	8.18	18.7	0.350	0.056	0.13	1.01 (0.23)	20	15.4	31.1 (1.0)	8.02 (3.3)
CE	7.94	503	8.22	18.8	–	–	–	0.06	757	7.04	14.9	0.110	0.050	0.18	4.85 (0.45)	22	15.7	21.5 (0.4)	34.6 (3.9)
OF	7.94	500	8.28	19.6	–	–	–	6.23	502	8.05	18.8	0.090	0.036	0.13	0.08 (0.04)	26	38.8	12.1 (0.5)	0.25 (0.3)
2018-June																			
IF	11.8	529	9.87	20.0	0.10	0.033	0.22	1.36	583	8.22	15.3	0.11	0.042	0.25	2.62 (1.02)	22	10.5	29.6 (2.2)	18.5 (7.3)
CE	11.8	528	9.14	19.4	0.11	0.045	0.16	0.60	764	8.30	10.4	0.12	0.055	0.24	3.10 (0.47)	22	14.7	22.0 (0.4)	18.2 (2.8)
OF	9.71	536	8.17	18.9	0.10	0.041	0.19	3.56	564	8.22	17.2	0.09	0.036	0.20	0.27 (0.05)	27	27.0	13.2 (0.2)	1.51 (0.3)
2018-Aug.																			
IF	13.8	346	8.71	20.3	0.28	0.14	0.15	0.27	799	7.07	13.8	0.28	0.190	0.23	5.47 (1.33)	20	11.8	24.7 (1.4)	35.6 (9.0)
CE	13.8	347	8.70	20.3	0.19	0.10	0.15	0.46	859	7.06	13.1	0.24	0.140	0.33	4.97 (0.48)	20	15.4	22.0 (0.2)	30.0 (3.0)
OF	10.8	362	8.47	20.4	0.16	0.093	0.13	1.48	443	7.68	19.9	0.16	0.091	0.14	0.31 (0.03)	20	24.1	12.8 (1.1)	1.40 (0.2)
2018-Oct.																			
IF	9.87	405	8.05	9.57	0.12	0.075	0.22	9.70	582	8.04	9.34	0.10	0.086	0.22	2.70 (2.05)	21	9.1	30.3 (2.5)	15.3 (12)
CE	11.0	411	8.19	10.7	0.11	0.079	0.20	8.66	573	7.82	9.54	0.10	0.066	0.22	2.17 (0.92)	20	14.3	23.5 (0.1)	12.8 (5.6)
OF	10.0	404	7.88	10.7	0.10	0.067	0.17	9.03	556	7.83	9.96	0.11	0.062	0.17	0.06 (0.01)	26	26.3	12.8 (1.4)	-0.03 (0.04)
1^{st} vs. 2^{nd} yr.																			
2017	8.1 b	501 a	8.3	19.3	–	–	–	4.7 a	588 a	7.8	17.5 a	0.18 a	0.05 a	0.15 a	2.0 a	23 a	23 a	22 a	14.3 a
2018	12.8	352 b	8.6	20.3	0.21	0.11	0.14	0.7 a	700 a	7.3	15.6 a	0.23 a	0.14 a	0.23 a	3.6 a	20 a	17 a	20 a	22.3 a
Seasonal																			
June	11.1 b	531 a	9.1	19.4	0.10 b	0.04 b	0.19a	1.8 b	637 a	8.2	14.3 a	0.11 b	0.04 b	0.23 a	2.0 a	24 a	17 a	22 a	12.7 ab
Aug.	12.8 a	352 c	8.6	20.3	0.21 a	0.11 a	0.14a	0.7 b	700 a	7.3	15.6 a	0.23 a	0.14 a	0.23 a	3.6 a	20 a	17 a	20 a	22.3 a
Oct.	10.3 b	407 b	8.0	10.3 b	0.11 b	0.07 b	0.20a	9.1 a	570 a	7.9	9.6 a	0.10 b	0.07 b	0.20 a	1.6 a	22 a	17 a	22 a	9.4 b
Spatial																			
IF	11.1	424 a	8.5	20.0	0.17	0.08 a	0.20a	4.1 a	652 ab	7.6	16.3 a	0.32 a	0.12 a	0.18 a	3.2 a	20 b	14 b	28 a	21.8 a

CE	10.9	425 a	8.5	19.6	0.14	0.07 a	0.17a	0.3 a	808 a	7.1	14.0	0.18 a	0.10 a	0.26 a	4.9 a	21 b	16 b	22 b	32.3 a
OF	9.4 b	431 a	8.4	20.0	0.12	0.07 a	0.16a	3.9 a	473 b	7.9	19.0	0.13	0.06 a	0.14	0.2 b	24 a	31 a	12 c	0.8 b

***** Mean values followed by a different letter are significantly different at $P < 0.05$.

Highlights

- We studied sediment P mobility in a small shallow lake after sediment excavation.
- After sediment removal, there was still high rate of summer P release from sediment.
- High external nutrient loading hindered improvements in water quality.
- Internal P loading sustained eutrophication of the lake.
- Sediment Fe-P and labile P fractions were the most important sources of P release.

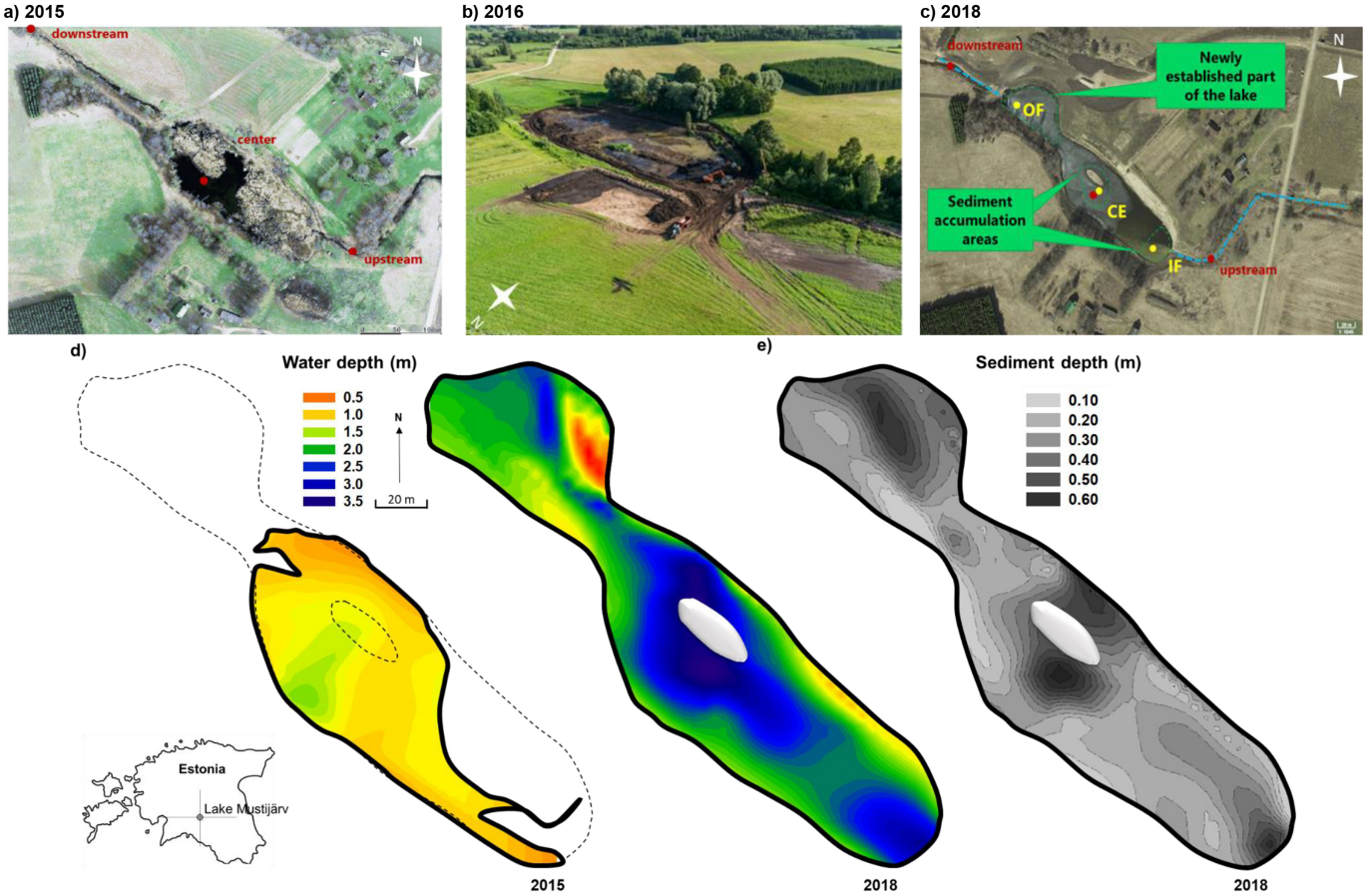
a) 2015
downstream
center
upstream
N
b) 2016
c) 2018
downstream
Newly established part of the lake
OF
CE
IF
upstream
Sediment accumulation areas
N
d)
Water depth (m)
0.5
1.0
1.5
2.0
2.5
3.0
3.5
N
20 m
Estonia
Lake Mustijärv
2015
e)
Sediment depth (m)
0.10
0.20
0.30
0.40
0.50
0.60
2018
2018

Figure 1

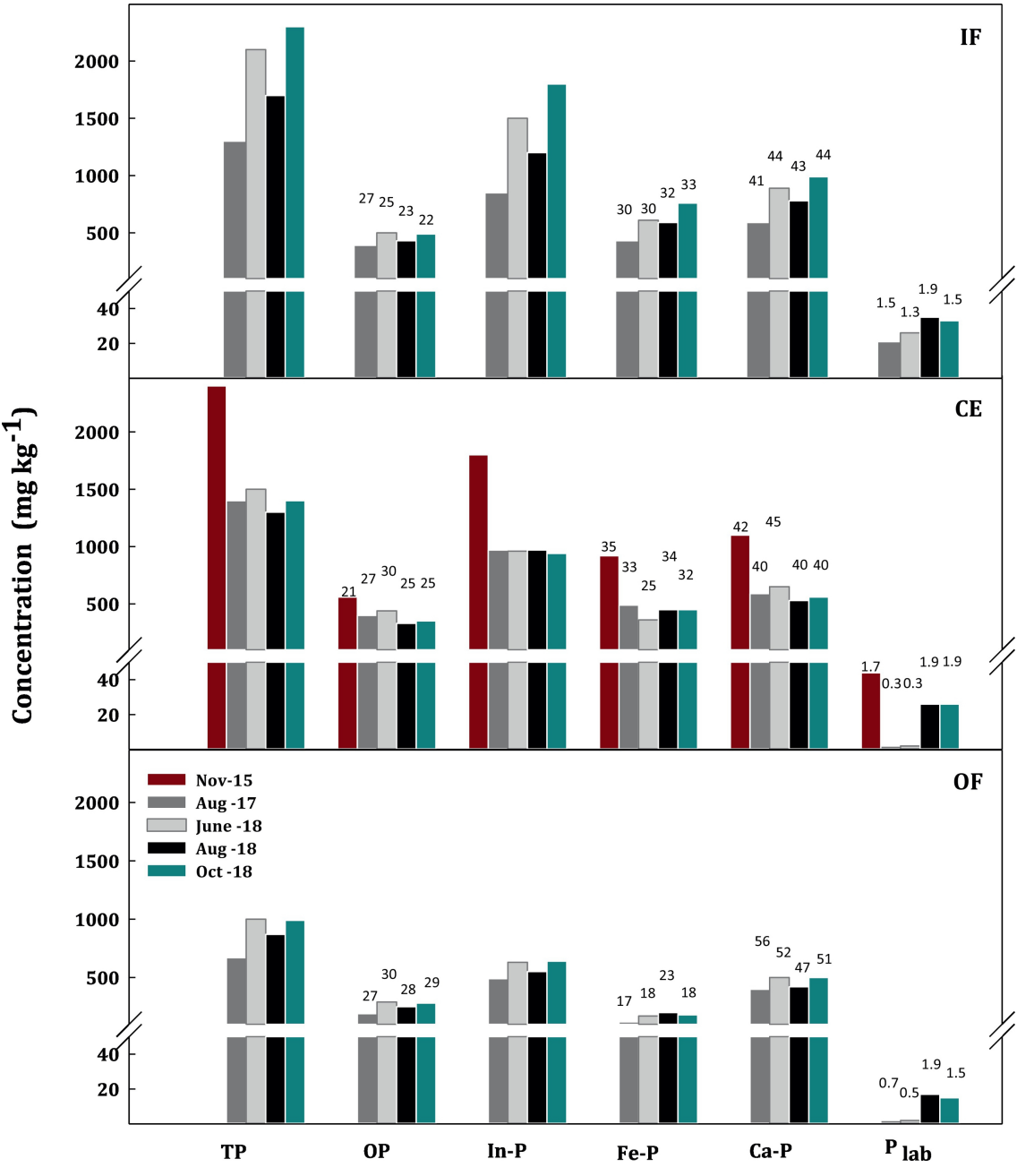
IF
CE
OF
Concentration (mg kg-1)
2000
1500
1000
500
40
20
27 25 23 22
30 30 32 33
41 44 43 44
1.5 1.3 1.9 1.5
21 27 30 25 25
35 33 25 34 32
42 40 45 40 40
1.7 0.3 0.3 1.9 1.9
27 30 28 29
17 18 23 18
56 52 47 51
0.7 0.5 1.9 1.5
Nov-15
Aug -17
June -18
Aug -18
Oct -18
TP
OP
In-P
Fe-P
Ca-P
P lab

Figure 2

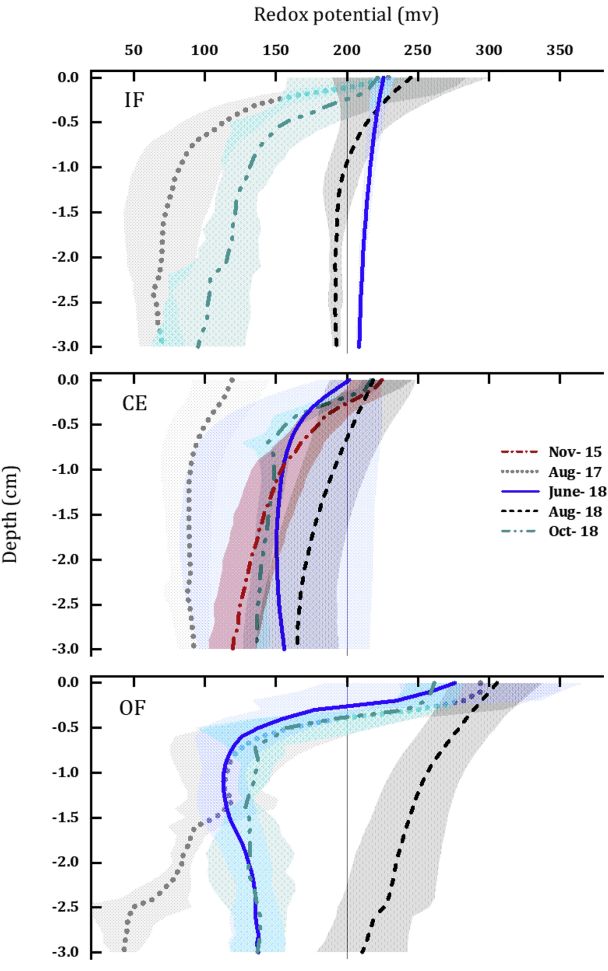
Redox potential (mv)
50
100
150
200
250
300
350
IF
CE
OF
Depth (cm)
0.0
-0.5
-1.0
-1.5
-2.0
-2.5
-3.0
Nov- 15
Aug- 17
June- 18
Aug- 18
Oct- 18

Figure 3

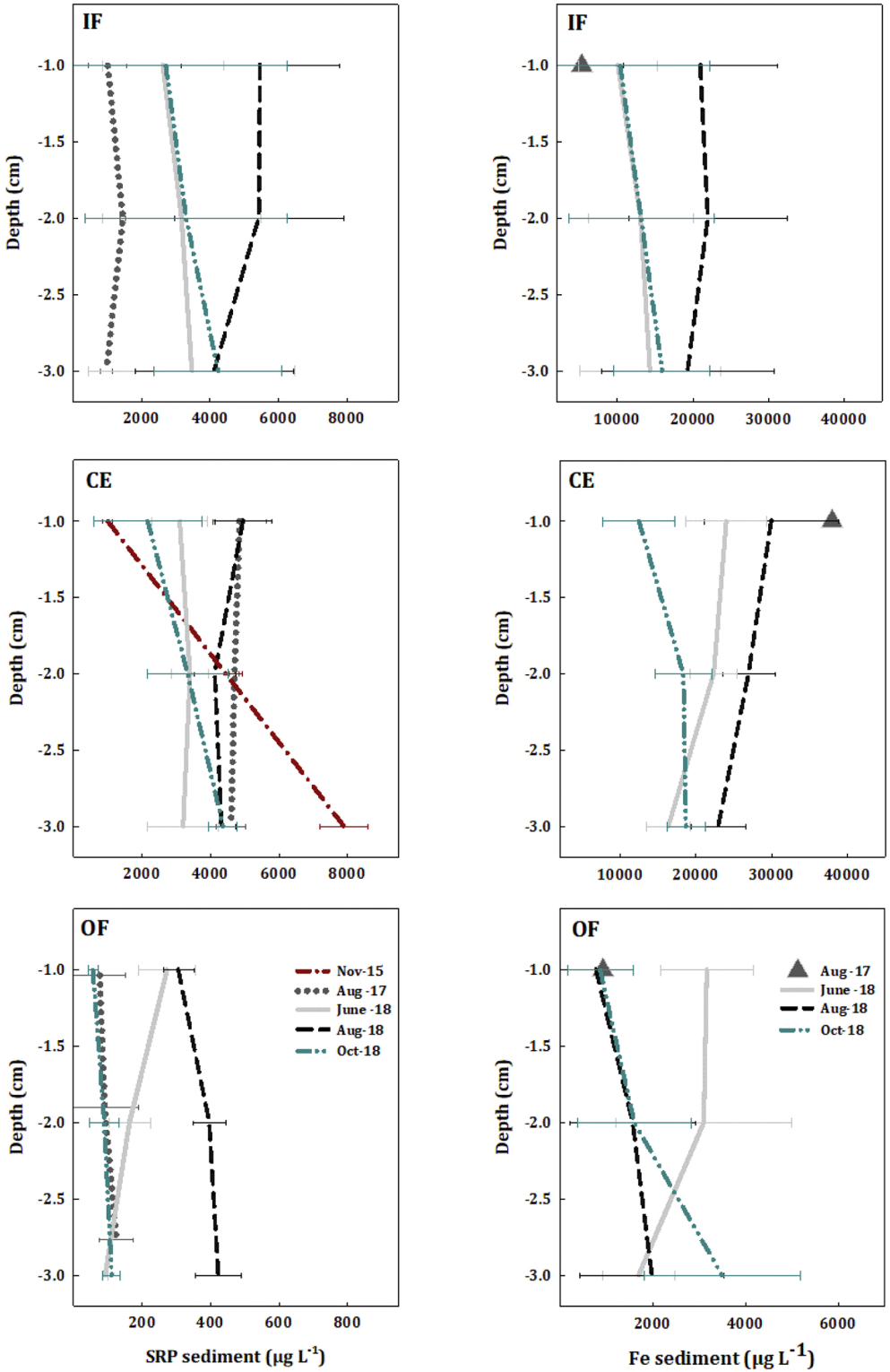
IF
IF
CE
CE
OF
OF
Depth (cm)
-1.0
-1.5
-2.0
-2.5
-3.0
2000
4000
6000
8000
10000
20000
30000
40000
200
400
600
800
Nov-15
Aug-17
June-18
Aug-18
Oct-18
SRP sediment (µg L-1)
Fe sediment (µg L-1)

Figure 4

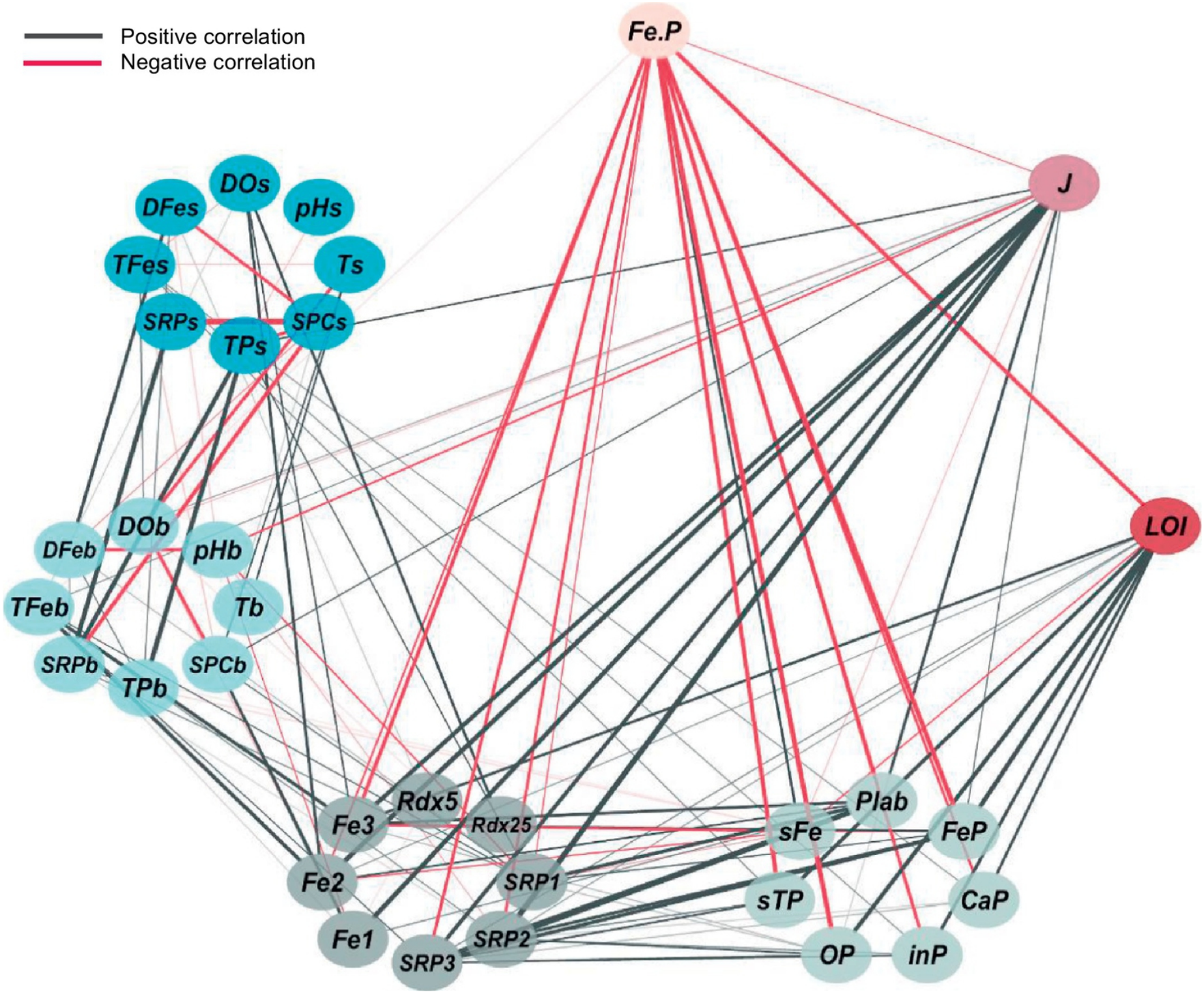
Positive correlation
Negative correlation
Fe.P
J
LOI
DOs
DFes
pHs
TFes
Ts
SRPs
SPCs
TPs
DOb
DFeb
pHb
TFeb
Tb
SRPb
SPCb
TPb
Rdx5
Fe3
Rdx25
Plab
sFe
FeP
Fe2
SRP1
sTP
CaP
Fe1
SRP2
SRP3
OP
inP

Figure 5